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⑤④ Self-extinguishing polymer composition.

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**EP-A- 0 212 575 EP-A- 0 221 449**  
**EP-A- 0 244 841 EP-A- 0 285 564**  
**FR-A- 2 357 569 GB-A- 1 372 538**  
**US-A- 3 580 735**

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## Description

The present invention relates to a self-extinguishing polymer composition, and particularly to a self-extinguishing polymer composition which is suitable for use in the fields of extrusion molded products such as wire coatings and pipes and injection molded products such as domestic electrical containers and electrical equipment connectors, and which has an extremely high degree of non-flammability and significantly improved practicability with respect to mechanical properties and acid resistance.

In recent years, the demand for imparting flame retardance to materials used for wire cables has significantly increased.

One example of a method of imparting flame retardance to thermoplastic resin or elastomers is a method in which antimony oxide and a halide are mixed in the raw materials used. Self-extinguishing compositions formed by this method, however, involve the danger of generating harmful gases during a fire. For this reason, hydrated metal compounds such as aluminum hydroxide or magnesium hydroxide or compounds thereof with magnesium carbonate have attracted attention because they generate no harmful gases and involve no problem with respect to toxicity as additives.

This method in which flame retardance is imparted to a thermoplastic resin or elastomer by compounding a hydrated metal compound therewith is already known. It is also known that flame retardance can be improved by adding a fine carbon powder to a composition containing a thermoplastic resin or elastomer and a hydrated metal compound (Japanese Patent Laid-Open No. 46341/1976).

Although the above-mentioned compound system is provided with a satisfactory level of flame retardance, there has been a problem in that it is very difficult to obtain a composition having a satisfactory level of practicability because whitening occurs in a resin composition having the above-described composition when any external force is applied thereto and because the resin composition exhibits a low level of acid resistance.

## SUMMARY OF THE INVENTION

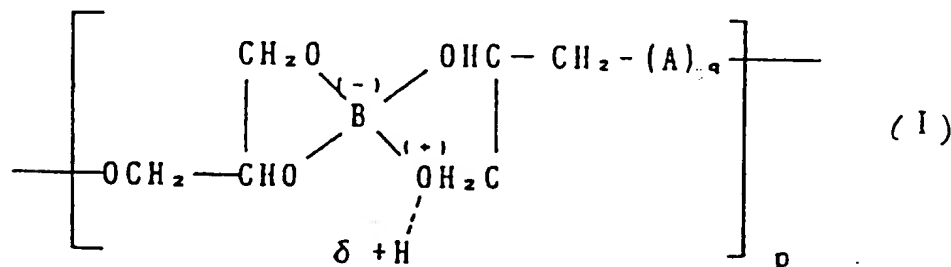
In the present invention, the aforementioned problem is resolved by compounding a boron compound with conventional inorganic flame retardants.

The present invention provides a self-extinguishing polymer composition consisting of 70 to 20% by weight of the component (A) described below, 30 to 80% by weight of the component (B) described below and the component (C) described below in an amount of 0.01 to 10 parts by weight relative to 100 parts by weight of the total amount of the components (A) and (B);

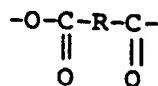
(A): one or more thermoplastic resins or elastomers derived from an  $\alpha$ -olefin polymer;

(B): a hydroxide of aluminum and/or magnesium;

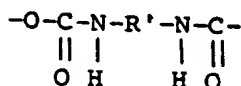
(C): a boron compound (polymeric charge-transfer type complex) which is a reaction product having a ratio of boron atoms to basic nitrogen atoms of 1 : 1 and is produced by reaction between at least one tertiary amine having at least one hydroxyl group and 5 to 82 carbon atoms in total and at least one semi-polar organic boron polymer compound expressed by the following Formula (I):



[wherein q denotes 0 or 1 and, when q = 1, A denotes a  $-(X)_r-(Y)_m-(Z)_n-$  group and p is 10 to 1000, in Formula (I), X and Z each denoting an oxygen-containing hydrocarbon group having one terminal ether residue and 100 or less carbon atoms in total, Y denoting a



group (wherein R represents a hydrocarbon group having 1 to 82 carbon atoms) or a



group (wherein R' represents a hydrocarbon group having 2 to 13 carbon atoms), l, m and n each denoting 0 or 1.]

#### DETAILED DESCRIPTION OF THE INVENTION

##### (A) Thermoplastic resins or elastomers

Examples of thermoplastic resins or elastomers that may be used as the above-described component (A) in the present invention include olefin polymers such as homopolymers of  $\alpha$ -olefins such as ethylene, propylene, butene, methylpentene, hexene and the like; copolymers of these olefins; and copolymers of these  $\alpha$ -olefins with 50% by weight or less of other comonomers.

Of these polymers, resins or elastomers of ethylene- or propylene-type polymers such as polyethylene, polypropylene, ethylene-propylene copolymer rubber, ethylene-propylene-diene copolymer rubber, ethylene-propylene copolymer resins, ethylene-vinyl acetate copolymer resins, ethylene-methyl acrylate resins, ethylene-methyl methacrylate copolymer resins, ethylene-ethyl acrylate resins are particularly preferable from the viewpoints of their molding properties and uniform dispersibility.

Of various olefinic polymer resins, linear ethylene polymers with density of 0.850 to 0.915 g/cm<sup>3</sup>, ethylene polymers containing 1 to 40% by weight of vinyl acetate and having melt flow rates (MFR) of 0.05 to 50 g/10 minutes and ethylene polymers containing 1 to 40% by weight of alkyl acrylate or alkyl methacrylate and having MFR of 0.05 to 50 g/10 minutes are particularly preferable from the viewpoint of remarkableness of the effect of the invention.

##### (B) Hydrated metal compound

The hydrated metal compound used as component (B) in the present invention is aluminum hydroxide and/or magnesium hydroxide. As occasion demands, a compound containing these metal compounds and 30 % by weight or less, preferably 5 to 20% by weight, of magnesium carbonate may be used. The average particle size of each metal compound is generally 0.1 to 20  $\mu\text{m}$ , preferably 0.2 to 2  $\mu\text{m}$ , and an average particle size of 0.5 to 1  $\mu\text{m}$  is particularly preferable from the viewpoint of dispersibility. Metal compounds which are subjected to surface treatment may be used.

Of these metal compounds, magnesium hydroxide is particularly preferable from the viewpoint of practicability.

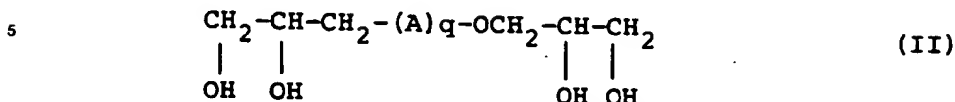
In addition, the use of magnesium hydroxide which was subjected to surface treatment using stearic acid or a metal salt of stearic acid is preferable because it enables the achievement of molded products exhibiting excellent physical properties and low levels of bleeding of magnesium carbonate, as well as having good appearance.

##### (C) Boron compound (polymeric charge-transfer type complex)

The particular boron compound (polymeric charge-transfer type complex) used as component (C) in the present invention is produced by the following reaction:

At least one tertiary amine having at least one hydroxyl group and 5 to 82 carbon atoms in total (referred to as "a given tertiary amine" hereinafter) is reacted with at least one semi-polar organic boron polymer compound (referred to as "a given semi-polar organic boron polymer compound" hereinafter) obtained by the following reaction (a), (b) or (c):

(a) Triesterification reaction between 1 mole of the total of at least one compound expressed by the following Formula (II):



[wherein q denotes 0 or 1 and, when q = 1, A denotes a  $-(\text{X})_r(\text{Y})_m(\text{Z})_n$  group, wherein X and Z each denoting an oxygen-containing hydrocarbon group having one terminal ether residue and 100 or less carbon atoms in total, Y denoting a



group (wherein R denotes a hydrocarbon group having 1 to 82 carbon atoms) or a



group (wherein R' denotes a hydrocarbon group having 2 to 13 carbon atoms) and l, m, n each denoting 0 or 1]

and 1 mole of boric acid or a triborate of a lower alcohol having 4 or less carbon atoms or 0.5 mole of boric anhydride;

(b) Polyetherification reaction using at least one di(glycerin) borate or diol having a di(glycerin) borate residue at an intermediate position in its molecule and 206 or less carbon atoms in total;

(c) Reaction between 1 mole of the total of at least one di(glycerin) borate or diol having a di(glycerin) borate residue at an intermediate position and 206 or less carbon atoms in total in its molecule and 1 mole of the total of at least one dicarboxylic acid having 3 to 48 carbon atoms (referred to as "a given dicarboxylic acid" hereinafter), an ester of a lower alcohol having 4 or less carbon atoms and a given dicarboxylic acid and a halide of a given dicarboxylic acid or a diisocyanate having 4 to 15 carbon atoms (referred to as "a given diisocyanate" hereinafter). The reaction between "a given tertiary amine" and "a given semi-polar organic boron polymer compound" is carried out wherein the materials are charged in a closed-type or open-type reactor so that a ratio of boron atoms to basic nitrogen atoms is 1 : 1 at a temperature of 20 to 200 °C, preferably 50 to 150 °C, and under a normal pressure. The reaction proceeds more easily if a polar solvent such as an alcohol, ether, ketone or the like is caused to coexist with the materials.

Examples of compounds expressed by Formula II which may be used as raw materials of the reaction (a) include diglycerin, di(glycerin) malonate, di(glycerin) maleate, di(glycerin) adipate, di(glycerin) terephthalate, di(glycerin) dodecanate, poly(9 moles)-oxyethylene di(glycerin) ether, di(glycerin) tolylene dicarbamate, di(glycerin) methylene bis(4-phenylcarbamate) and the like.

Examples of given dicarboxylic acids which may be used in the reaction (c) include malonic acid, maleic acid, succinic acid, adipic acid, sebacic acid, phthalic acid, terephthalic acid, dodecanedicarboxylic acid, dimer acid derived from linolic acid, dodecyl maleate, dodecenyl maleate, octadecyl maleate, octadecenyl maleate, maleic acid to which a polybutenyl group with an average degree of polymerization of 20 is jointed and the like.

Examples of given diisocyanates include ethylene diisocyanate, hexamethylene diisocyanate, tolylene diisocyanate, methylene bis(4-phenylisocyanate) and the like.

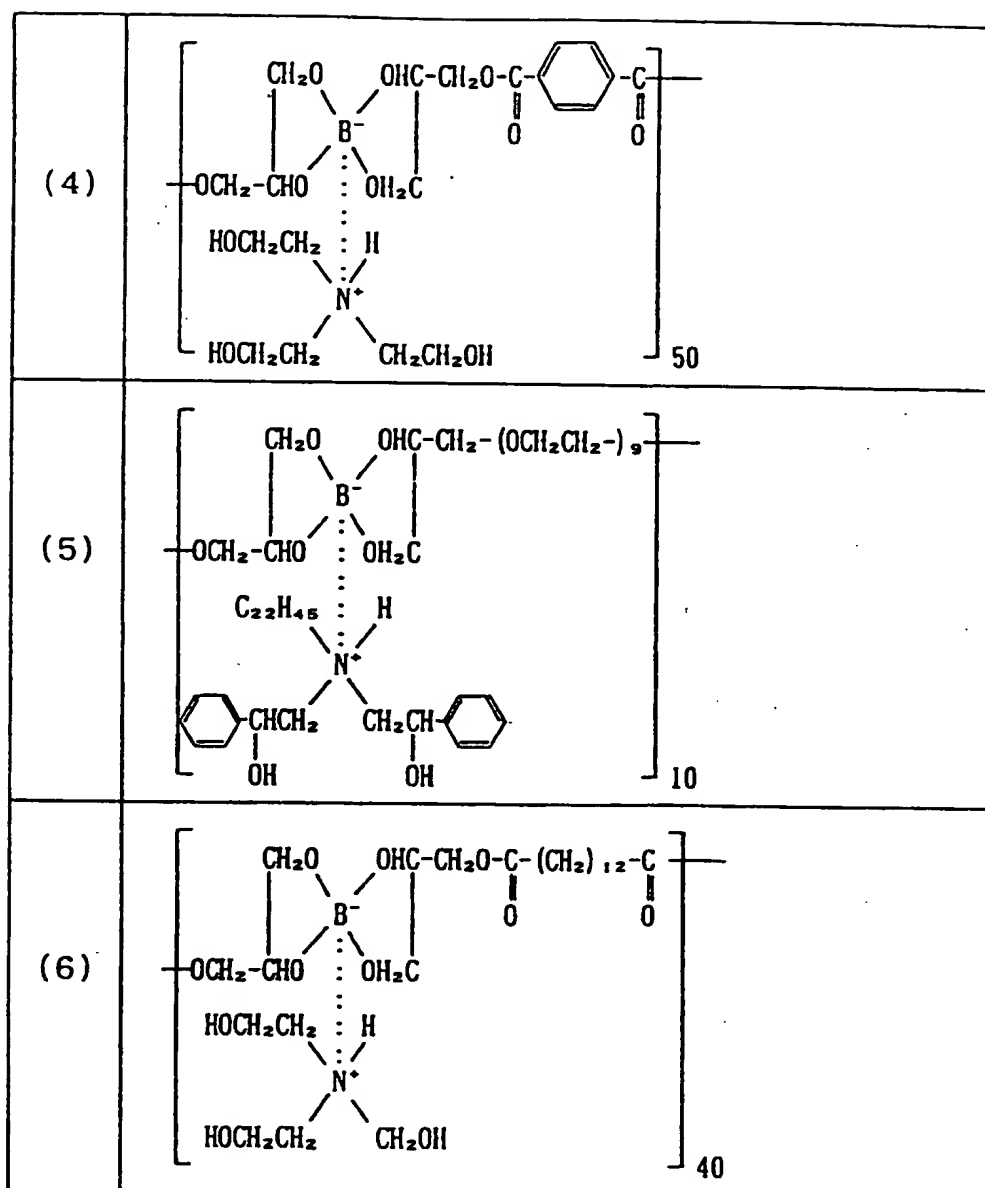
Examples of given tertiary amines which may be reacted with given semi-polar organic boron polymer compounds include diethyl hydroxymethylamine, dimethyl 2-hydroxypropylamine, methyl di(2-hydroxyethyl)amine, tri(2-hydroxyethyl)amine, hydroxymethyl di(2-hydroxyethyl)amine, dibenzyl 2-hydroxypropylamine, cyclohexyl di(2-hydroxyethyl)amine, ethylene oxide (1 to 25 moles) addition products of di(hexadecyl)amine, propylene oxide (1 to 26 moles) addition products of monobutylamine and the like.

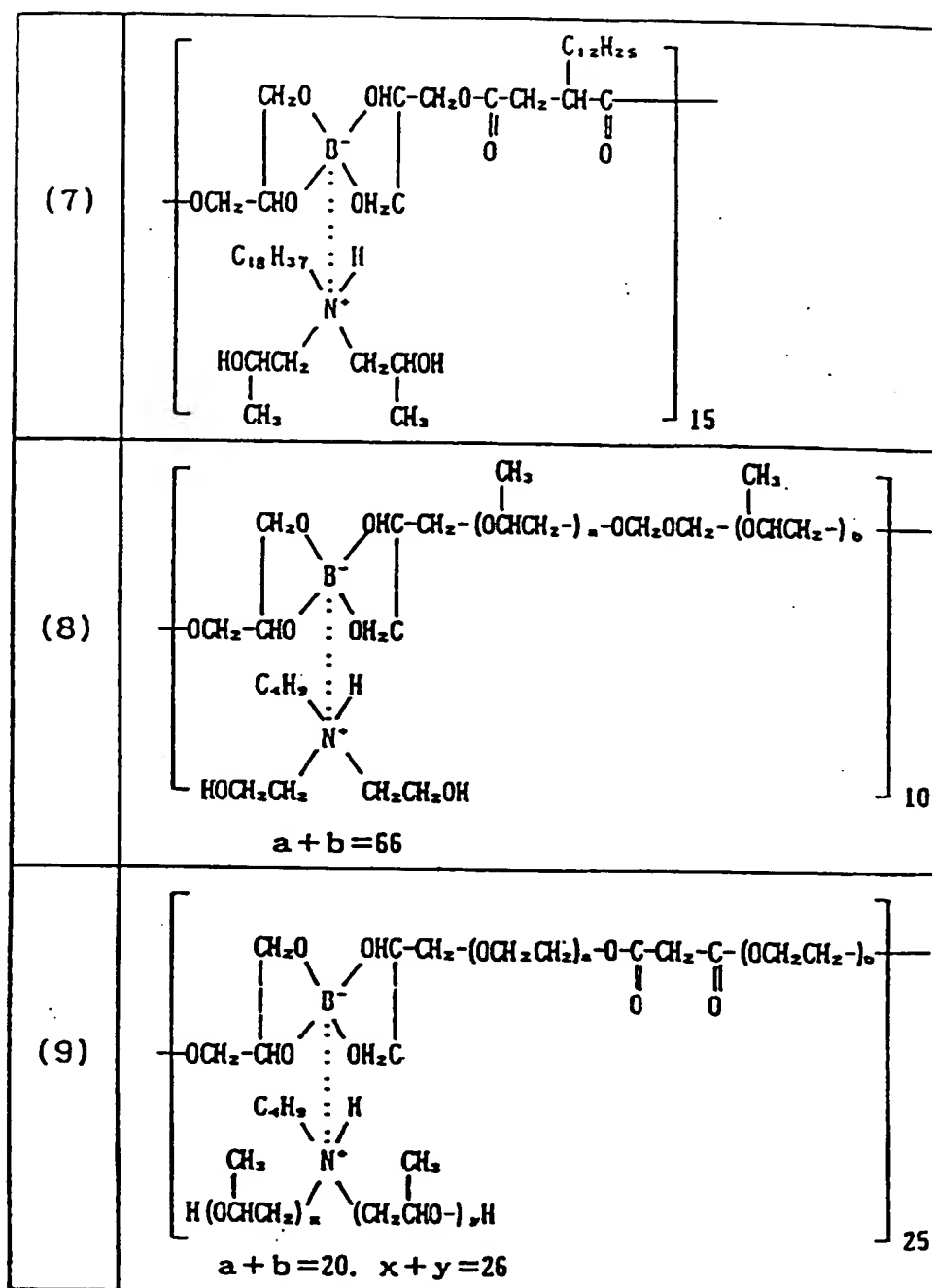
Typical examples of boron compounds (polymeric charge-transfer type complex) that may be used in the present invention are listed in Table 1.

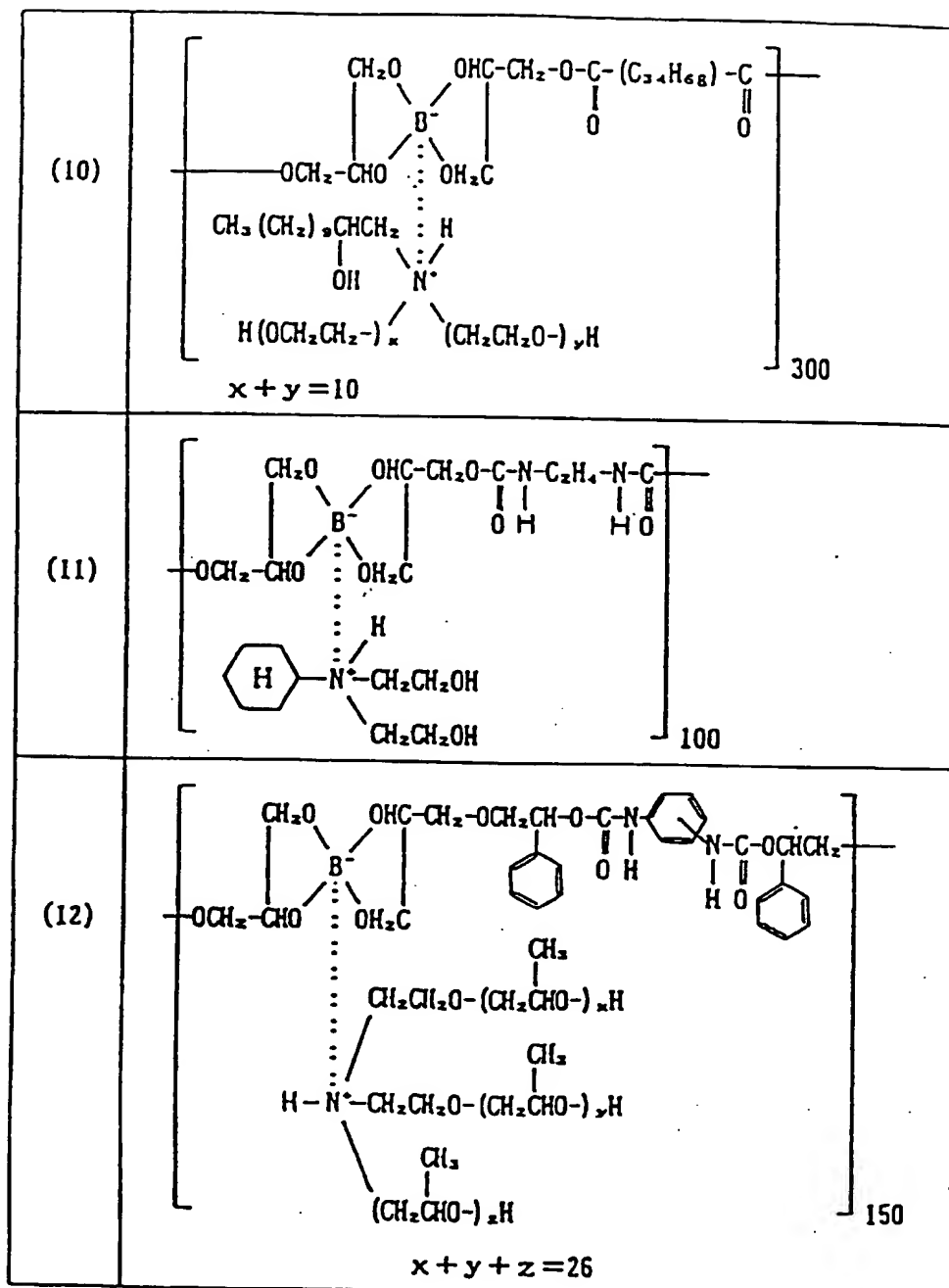
With respect to the compounding ratio of the necessary components used in the present invention, the proportion of the component (A) is 70 to 20% by weight, preferably 65 to 35% by weight, of the total amount of the components (A) and (B), and the proportion of the necessary component (C) is 0.01 to 10 parts by weight, preferably 0.05 to 5 parts by weight, more preferably 0.05 to 2 parts by weight, relative to 100 parts by weight of the total amount of the components (A) and (B). If the proportion of the component (C) is too small, resistance to mechanical whitening and acid resistance cannot be improved, while, if the proportion is too large, molding properties significantly deteriorate.

Table 1

Typical Example	Structural Formula
(1)	
(2)	<p>R denotes a polybutene residue having an average polymerization degree of 20.</p>
(3)	











composition exhibiting a high level of non-flammability and good resistance to mechanical whitening and acid resistance.

The composition of the present invention can be produced by compounding the aforementioned necessary components (A), (B) and (C) and, if required, additional components and then kneading the thus-formed compound by a normal kneading method such as an extruder, roll, Banbury mixer or the like. During the production, although any desired compounding order can be employed, it is preferable to use a method in which all the components are mixed in a dry blending manner and then kneaded. It is particularly preferable to use a method in which the component (B) is subjected to surface treatment using the component (C), then mixed with the other components in a dry blending manner and then kneaded.

Although molding is generally performed by using pellets of the composition of the present invention, molding can also be performed after mixing or during mixing where a master batch or dry blending method is adapted.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

##### (EXAMPLES)

The boron compound used in the examples described below is Typical Example (1) shown in Table 1 and produced by the following method:

##### Production of Boron Compound

1 mole of di(glycerin) borate and 1 mole of adipic acid were charged in a four-necked flask provided with a stirrer, a thermometer, a nitrogen gas inlet tube and a water measuring tube, and reaction was effected under a stream of nitrogen gas at 220 °C to 230 °C for 4 hours until 2 moles of water was removed. After an increase in the melt-viscosity had been recognized, the reaction product was cooled to 70 °C, and methyl ethyl ketone was charged in an amount which was substantially the same as that of the reaction product to form a uniform solution. 1 mole of poly(25 moles)-oxyethylene dihexadecylamine was then charged to the resultant solution, followed by reaction at 70 to 75 °C for 1 hour. After the reaction, methyl ethyl ketone used as a diluting solvent was distilled off to the outside of the system at 120 to 130 °C over the time of 2 hours under a reduced pressure of 150mmHg to obtain the boron compound (1).

##### Examples 1 to 14 and Comparative Examples 1 to 3

Each of the compositions having the compounding ratios shown in Table 2 was produced by kneading at 160 °C for 20 minutes using a laboratory plastomill.

The oxygen index (LOI) of each of the compositions obtained was evaluated in accordance with the method of JIS K7201, and the tensile strength and tensile elongation were evaluated in accordance with JIS K6760. The resistance to whitening resistance to acid and bleeding were evaluated by the following method:

##### (Whitening)

A sheet having a thickness of 1 mm was bent at an angle of 120 degrees, and the appearance of the sheet was visually observed. The decision of evaluation was made using the following criteria:

excellent:	no whitening or substantially no whitening
good:	a low degree of whitening
poor:	a high degree of whitening

##### (Acid resistance)

A sheet having a thickness of 1 mm was soaked in a 10% hydrochloric acid at 50 °C for 72 hours, and the retention rate of weight was then measured.

##### (Bleeding)

A sheet having a thickness of 1 mm was allowed to stand in an atmosphere of water vapor saturated with carbon dioxide gas at room temperature for 3 days and then dried. The appearance of the sheet was then visually observed. The decision of evaluation was made by using the following criteria:

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excellent: no bleeding or substantially no bleeding of magnesium carbonat or aluminum carbonate  
 good: a low degree of bleeding  
 poor: a high degree of bleeding

The abbreviations of components shown in Table 2 respectively represent the following substances:

- 5 PE: Polyethylene having a MFR value of 1.0 g/10 minutes and a density of 0.90 g/cm<sup>3</sup>  
 EMA: Ethylene-ethyl acrylate polymer having a MFR value of 2.0 g/10 minutes and containing 18% by weight of ethyl acrylate  
 Mg(OH)<sub>2</sub> [I]: Mg(OH)<sub>2</sub> (average particle size, 0.6 μm) subjected to surface treatment with 1 part by weight of the boron compound relative to 100 parts by weight of Mg(OH)<sub>2</sub>  
 10 Mg(OH)<sub>2</sub> [II]: Mg(OH)<sub>2</sub> (average particle size, 0.6 μm) subjected to surface treatment with 1 part by weight of the boron compound and 1 part by weight of sodium stearate relative to 100 parts by weight of Mg(OH)<sub>2</sub>  
 Mg(OH)<sub>2</sub> [III]: Mg(OH)<sub>2</sub> (average particle size, 0.6 μm) subjected to surface treatment with 0.5 part by weight of the boron compound and 1 part by weight of sodium stearate relative to 100 parts by weight of  
 15 Mg(OH)<sub>2</sub>  
 Mg(OH)<sub>2</sub> [IV]: Mg(OH)<sub>2</sub> (average particle size, 0.6 μm) subjected to surface treatment with 0.1 part by weight of the boron compound and 1 part by weight of sodium stearate relative to 100 parts by weight of Mg(OH)<sub>2</sub>  
 Mg(OH)<sub>2</sub> [V]: Mg(OH)<sub>2</sub> having an average particle size of 0.6 μm and subjected to no surface treatment  
 20 Mg(OH)<sub>2</sub> [VI]: Mg(OH)<sub>2</sub> (average particle size, 0.6 μm) subjected to surface treatment with 1 part by weight of sodium stearate relative to 100 parts by weight of Mg(OH)<sub>2</sub>  
 Al(OH)<sub>3</sub>: aluminium hydroxide having an average particle size of 0.6 μm and subjected to no surface treatment  
 Carbon: Carbon black having an average particle size of 19 μm and a specific surface area of 140 m<sup>2</sup>/g  
 25 Silicon: Silicon diol having viscosity of 55 cp, a refractive index of 1.41 and a specific gravity of 0.96  
 PbSt: Lead stearate

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Table 2

Ex- am- ple	Compara- tive Ex- am- ple	Compound Composition											LOI	Whitening	Acid resist- ance %	Tensile strength kg/mm <sup>2</sup>	Tensile elonga- tion %	Blood- ing
		Polymer		Hydrated metal compound		Carbon compound content	Silicone compound		PBst Con- tent									
		Type	Content	Type	Content		Type	Content										
1		PE	47	Mg(OH) <sub>2</sub> (II)	53	(0.53)*	-	-	-	-	28	good	90	100	650	good		
2		"	"	Mg(OH) <sub>2</sub> (III)	"	"	-	-	-	-	28	good	97	100	650	excellent		
3		PMA	"	"	"	"	-	-	-	-	28	good	97	100	650	excellent		
4		PE	"	"	"	"	2.5	-	-	-	28	excellent	97	100	650	excellent		
5		"	"	"	"	"	-	-	-	-	28	good	97	120	700	excellent		
6		"	"	"	"	"	2.5	-	-	-	38	excellent	97	120	700	excellent		
7		"	"	"	"	"	-	-	-	0.5	42	excellent	97	130	700	excellent		
8		"	40	"	60	"	-	-	-	-	45	good	90	120	600	good		
9		"	"	Mg(OH) <sub>2</sub> (III)	"	(0.30)*	-	-	-	-	45	good	90	120	600	good		
10		"	"	Mg(OH) <sub>2</sub> (IV)	"	(0.06)*	-	-	-	-	45	good	70	120	600	good		
11		"	"	Al(OH) <sub>3</sub>	"	0.50	-	-	-	-	42	good	90	110	600	excellent		
12		"	47	Mg(OH) <sub>2</sub> (IV)	53	"	-	-	-	-	28	good	60	100	650	good		
13		"	"	"	"	"	2.5	-	-	-	0.5	42	excellent	60	130	700	excellent	
14		"	"	Mg(OH) <sub>2</sub> (VII)	"	"	-	-	-	-	42	excellent	60	130	700	excellent		
1		"	"	"	"	"	-	-	-	-	28	poor	50	100	650	good		
2		"	"	"	"	"	2.5	-	-	-	0.5	42	good	50	130	700	good	
3		"	40	"	60	"	-	-	-	-	45	poor	45	120	600	good		
4		"	47	Mg(OH) <sub>2</sub> (IV)	53	"	-	-	-	-	28	poor	50	100	650	poor		

\* The numerical values in each parenthesis represents the amount used for surface treatment of a hydrated metal compound.

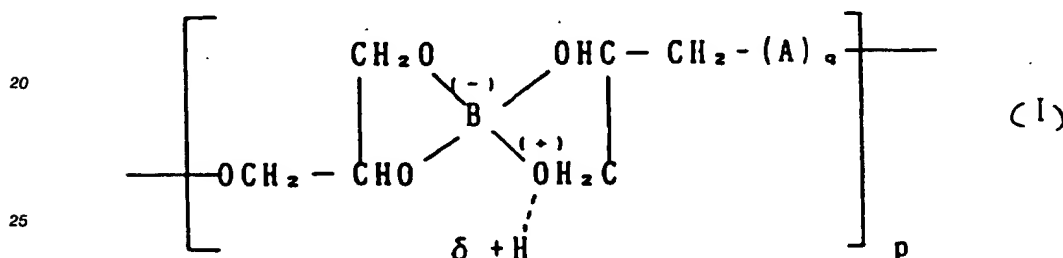
Content: in terms of parts by weight

As can be seen from Table 2, the compositions of Examples 1 to 14 which contained hydrated metal salts subjected to surface treatment with the boron compound exhibit excellent properties with respect to resistance to whitening and acid resistance, as compared with the compositions of Comparative Examples 1 to 3 which contained no boron compound.

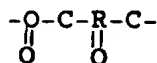
In the present invention, the use of a particular amount of a particular boron compound together with hydrated metal compounds of the type which is generally mixed in conventional flame retardant compositions enables the achievement of a self-extinguishing composition exhibiting significantly improved properties such as nonflammability, tensile strength, tensile elongation, control of whitening, acid resistance and so forth. Thus, the practical performance which has been a problem of conventional composition is significantly improved.

# Claims

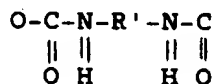
1. A self-extinguishing polymer composition comprising (A) from 70 to 20% by weight of one or more thermoplastic resins or elastomers derived from an  $\alpha$ -olefin polymer; and (B) from 30 to 80% by weight of a hydroxide of aluminium and/or magnesium; together with (C) from 0.01 to 10 parts by weight, per 100 parts by weight of the total of components (A) and (B); of a polymeric charge-transfer type complex which is a reaction product having a ratio of boron atoms to basic nitrogen atoms of 1:1 and is produced by reaction between at least one tertiary amine having at least one hydroxyl group and a total of 5 to 82 carbon atoms and at least one semi-polar organic boron polymer compound of the formula:



(wherein  $p$  is 10 to 1000,  $q$  is 0 or 1 and, when  $q = 1$ , A is a group  $-(X)_l-(Y)_m-(Z)_n-$  in which X and Z are each an oxygen-containing hydrocarbon group having one a terminal ether residue and a total of 100 or less carbon atoms; Y is a group



group  
(wherein R is a  $\text{C}_1$ - $\text{C}_{82}$  hydrocarbon group) or a group

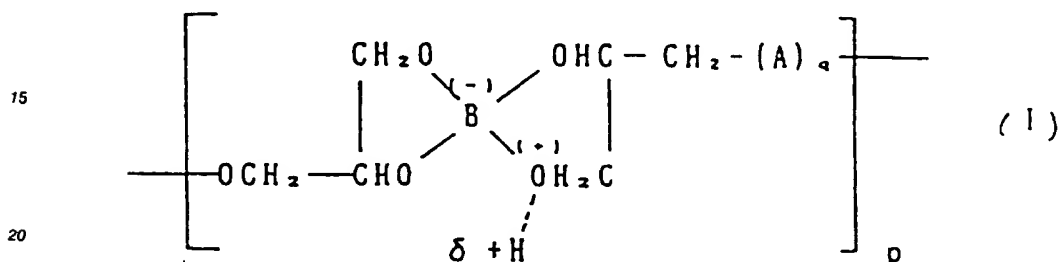


(wherein  $\text{R}'$  represents a  $\text{C}_2$ - $\text{C}_{13}$  hydrocarbon group); and  $l$ ,  $m$  and  $n$  are each 0 or 1].

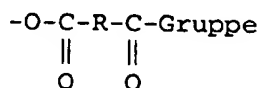
2. A composition according to claim 1, containing from 35 to 65% by weight of component (A) relative to the total amount of components (A) and (B).
3. A composition according to claim 1 or claim 2 containing 0.05 to 5 parts by weight of component (C), per 100 parts by weight of the total of components (A) and (B).
4. A composition according to any one of the preceding claims in which the thermoplastic resin is polyethylene or an ethylene-acrylic ester copolymer.

## Patentanspruch

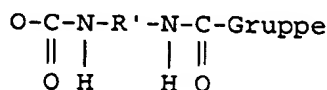
1. Selbstverlöschende Polymerzusammensetzung, umfassend (A) 70 bis 20 Gew.-% eines oder mehrerer von einem  $\alpha$ -Olefinpolymer abgeleiteten thermoplastischen Harze oder Elastomere; und (B) 30 bis 80 Gew.-% eines Hydroxids des Aluminiums und/oder Magnesiums; zusammen mit (C) 0,01 bis 10 Gew.-Teile, pro 100 Gew.-Teile der gesamten Komponenten (A) und (B), eines polymeren Charge-Transfer-Komplexes, der ein Reaktionsprodukt mit einem Verhältnis von Boratomen zu basischen Stickstoffatomen von 1:1 ist und hergestellt wird durch Reaktion zwischen mindestens einem tertiären Amin mit mindestens einer Hydroxylgruppe und insgesamt 5 bis 82 Kohlenstoffatomen und mindestens einer semipolaren organischen Borpolymerverbindung der Formel



[worin p 10 bis 1000 ist, q 0 oder 1 ist, und wenn q = 1 ist, A eine Gruppe  $-(X)_l-(Y)_m-(Z)_n-$  ist, worin X und Z jeweils eine sauerstoffhaltige Kohlenwasserstoffgruppe mit einem terminalen Etherrest und insgesamt 100 oder weniger Kohlenstoffatomen sind; Y eine



(worin R eine  $\text{C}_1$ - $\text{C}_{82}$ -Kohlenwasserstoffgruppe ist) oder eine



ist

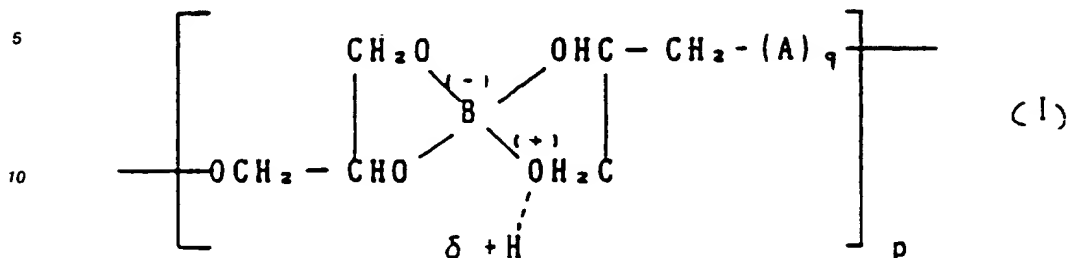
(worin R' eine  $\text{C}_2$ - $\text{C}_{13}$ -Kohlenwasserstoffgruppe darstellt); und l, m und n jeweils 0 oder 1 sind].

2. Zusammensetzung gemäß Anspruch 1, enthaltend 35 bis 65 Gew.-% Komponente (A), bezogen auf die Gesamtmenge der Komponenten (A) und (B).
3. Zusammensetzung gemäß Anspruch 1 oder 2, enthaltend 0,05 bis 5 Gew.-Teile Komponente (C) pro 100 Gew.-Teile der Gesamtmenge der Komponenten (A) und (B).
4. Zusammensetzung gemäß einem der vorhergehenden Ansprüche, worin das thermoplastische Harz Polyethylen oder ein Ethylen-Acrylester-Copolymer ist.

## Revendications

1. Composition polymère auto-extinguible comprenant (A) de 70 à 20 % en poids d'un ou plusieurs résine(s) thermoplastique(s) ou élastomère(s) dérivés d'un polymère de  $\alpha$ -oléfin(e)s ; et (B) de 30 à 80 % en poids d'un hydroxyde d'aluminium et/ou de magnésium ; ainsi que (C) de 0,01 à 10 partie(s) en poids, pour 100 parties en poids du total des composants (A) et (B), d'un complexe polymère de type à transfert de charge, qui est un produit réactionnel ayant un rapport des atomes de bore aux atomes d'azote basiques de 1:1 et qui est préparé par une réaction entre au moins une amine tertiaire ayant au

moins un groupe hydroxyle et un total de 5 à 82 atomes de carbone, et au moins un composé du bore polymère organique semi-polaire, de la formule :



[où  $p$  est 10 à 1 000,  $q$  est 0 ou 1 et, lorsque  $q = 1$ , A est un groupe  $-(X)_l-(Y)_m-(Z)_n$  où X et Z sont chacun un groupe hydrocarboné contenant de l'oxygène, ayant un reste d'éther terminal et un total de 100 atomes de carbone ou moins ; Y est un groupe



(où R est un groupe hydrocarboné en  $\text{C}_1$  à  $\text{C}_{28}$ ) ou un groupe



(où R' représente un groupe hydrocarboné en  $\text{C}_2$  à  $\text{C}_{13}$ ) ; et l, m et n sont chacun 0 ou 1].

2. Composition selon la revendication 1, contenant de 35 à 65 % en poids de composant (A) par rapport à la quantité totale des composants (A) et (B).

3. Composition selon la revendication 1 ou la revendication 2, contenant 0,05 à 5 partie(s) en poids de composant (C), pour 100 parties en poids du total des composants (A) et (B).

4. Composition selon l'une quelconque des revendications précédentes, dans laquelle la résine thermoplastique est du polyéthylène ou un copolymère éthylène-ester acrylique.